Irreversible elastic responses of single collapsed DNA molecules

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多価陽イオンで凝縮した一分子ＤＮＡの力学応答には伸張–緩和サイクルで履歴が生じる。力学応答の不可逆性を明らかにするため、伸張–緩和速度を変化させ不可逆仕事の速度依存性を調べた。0.1 – 2 μm/sec の範囲では不可逆仕事の大きさが速度に比例して増加するとともに、速度０で有限の仕事が残る結果が得られた。ＤＮＡ鎖の時間的、空間的緩和、及び凝縮の核形成過程がこれらの結果に深く関わっている。

DNA is a semiflexible polymer whose persistence length is about 50 nm, and highly negatively charged with two negative elementary charges per 0.34 nm. These characters play an important role in not only physical but also biological systems [1]. Multivalent cations induce the conformational change of a single DNA molecule from a wormlike coil state to a collapsed state, which is a well known phenomenon called DNA condensation [2]. Single collapsed DNA shows peculiar elastic responses depending on the cation concentrations [3]. Near the onset concentration of multivalent cations in the collapsing transition, force–extension (f–x) curves show force plateau in a wide range of the extension; the force plateau results from the coexistence of wormlike coil and collapsed states in the single DNA. Furthermore, the elastic responses of single collapsed DNA always show hysteresis on f–x curves during a stretch–relax (s–r) cycle. To elucidate the irreversibility, we have investigated the elastic responses during a s–r cycle of a single collapsed DNA using dual trap optical tweezers with varying the s–r speed systematically. We focused on the force plateau regime with fixing the concentration of trivalent cation, spermidine (SPD), at 400 μM which is near the onset concentration in the collapsing transition. We have found that the irreversible work ΔW during a s–r cycle increases as the s–r speed increases.

Both ends of λ-Phage DNA of 48.5 kilo-basepairs which corresponds to the contour length of 16.5 μm were attached to streptavidin-coated beads of 2.0 μm diameters, via biotinylated oligonucleotides that hybridized to the single-stranded ends of λDNA. Figure 1 shows typical f–x curves of a collapsed DNA during a s–r cycle at the speed of 1.26 μm/sec. In the presence

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of SPD, hysteresis on f–x curve always appeared during the cycle. We numerically calculated a irreversible work, $\Delta W = \int_{x_1}^{x_2}(f_{st} - f_{re})dx$, from the measurements, where x is the extension of the DNA, f_{st} and f_{re} are the force in stretching and relaxing, respectively. To produce the same conditions in different cycles, we waited for 2 min. at positions A and B in Fig. 1 during a cycle.

In case of collapsed DNA, ΔW linearly increases as the s–r speed increases, while no speed dependence appears for uncollapsed DNA (Fig. 2). Note that finite ΔW ($\equiv \Delta W_0$) remains at low speed limit. The finite ΔW_0 is an unexpected result, because in a quasi-static limit, a reversible elastic response should be obtained, *i.e.*, ΔW_0 must be zero. The f–x curves during a s–r cycle give us a hint to elucidate this unexpected fact. Note that the force starts to increase at some extension (x_c in Fig. 1) in the relaxing process. The increase of force in the relaxing process indicates the nucleation of the collapsing. In our measurements, we have found that no data shows $x_c \geq 8.0$ μm, moreover, the x_c tends to decrease as the speed increases; DNA is always in an uncollapsed state above the extension of 8.0 μm in the relaxing process at any speed. Thus, ΔW above the extension of 8.0 μm remains even at the lowest speed, which causes the finite ΔW_0. Brownian dynamics simulations of a single polyelectrolyte have qualitatively reproduced the speed dependence.

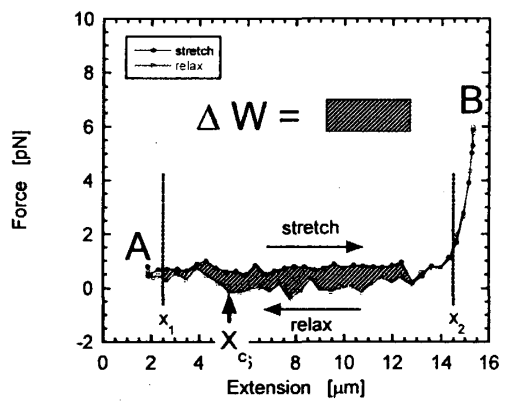

Figure 1: Typical f–x curves of a collapsed DNA in a s–r cycle at 400 μM SPD.

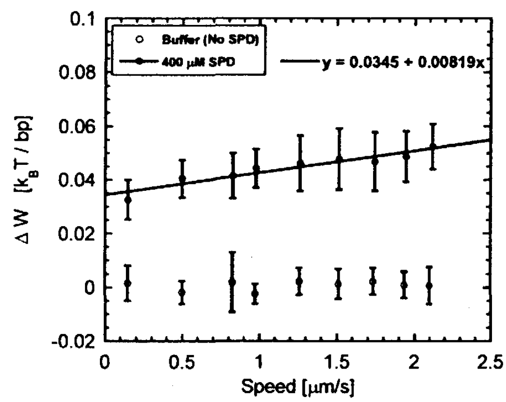

Figure 2: The dependence of ΔW on the s–r speed at 400 μM SPD (closed circle) and in buffer (open circle).

References

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